

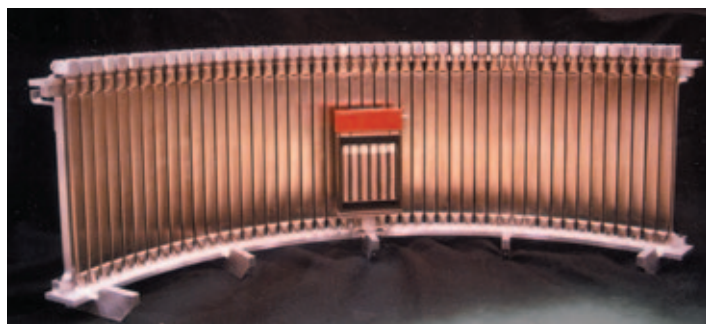
FILTER-ANALYZER NEUTRON SPECTROMETER (FANS)

Neutron vibrational spectroscopy (NVS) is an invaluable technique for probing vibrational dynamics. In particular, the typical range of energies accessible with reactor-based neutrons spans the region of important lattice and molecular vibrations. Moreover, the unique nature of the neutron-nucleus interaction permits the observation of all vibrational modes in an NVS experiment, not just those that satisfy appropriate symmetry-based selection rules as in photon spectroscopies. NVS is particularly useful for characterizing hydrogenous materials since the incoherent scattering cross section for hydrogen is much larger than for virtually all the other elements.

Over the past twenty years, neutron vibrational spectra have been collected for a broad array of both hydrogenous and non-hydrogenous systems utilizing a first-generation Filter-Analyzer Neutron Spectrometer (FANS) located at BT-4. Although this instrument possessed an excellent signal/noise ratio, the measured scattering intensities were limited by a relatively small detector solid angle. Moreover, the BT-4 beam time was typically shared with a triple-axis instrument. A second-generation FANS is currently being constructed at BT-4 in two phases in order to obtain a dedicated

FANS spectrometer with an enhanced measurement sensitivity of about two orders of magnitude. This instrument will ultimately overcome the classic objections to NVS, namely the requirement for much larger samples and longer counting times than are typical of infrared and Raman spectroscopies. Phase I of this second-generation instrument (FANS-I) is now complete, yielding a twenty-fold increase in sensitivity with another factor of at least five anticipated upon completion of phase II.

Figure 1 illustrates the FANS layout. Using either a Cu(220) or pyrolytic graphite (PG) (002) monochromator covers an accessible



Photography by L. A. Stuman

FIGURE 2. Comparison of the larger phase-I and original FANS detector banks.

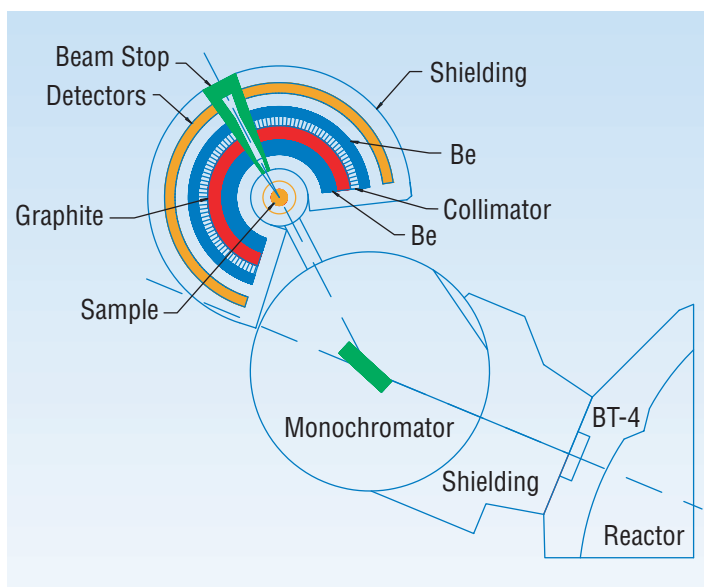


FIGURE 1. Schematic diagram of the final configuration of FANS. Presently installed phase I includes the filters to the right of the beam.

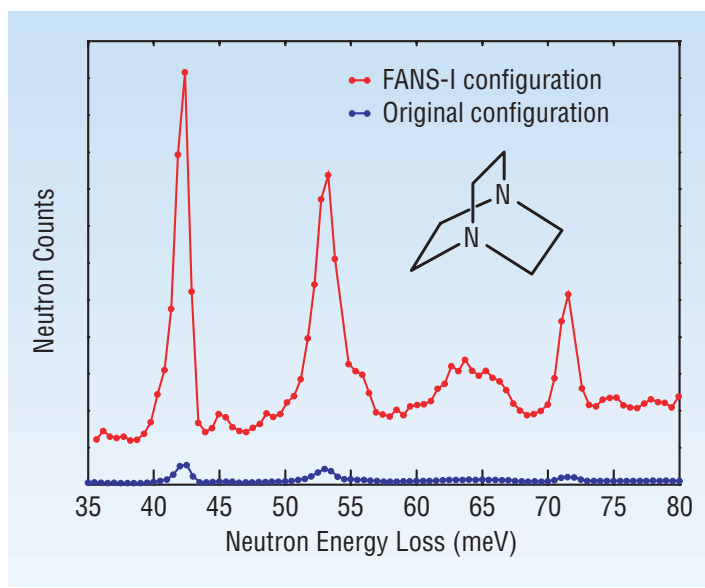


FIGURE 3. Comparison of the neutron vibrational spectrum for triethylene diamine ($C_2H_4)_3N_2$ measured with FANS-I vs. that measured with the original configuration.

energy range of ≈ 5 meV to 250 meV with options for precollimations of 60', 40', or 20' and postcollimations of 40', 20', or 10'. The intensity improvement of the phase-I instrument is due to the twenty-fold increase in detector solid angle provided by a much larger detector bank (see Fig. 2). The accompanying analyzer filter consists of a Be-PG-Be layered arrangement cooled with liquid nitrogen to enhance transmission of the low energy neutrons. Such a low-bandpass filter provides a best resolution at the lowest energy transfers of ≈ 1.1 meV FWHM.

The first measurements during the spring of 2000 confirmed the magnitude of the expected gains. For example, Fig. 3 displays the low-temperature neutron vibrational spectrum for triethylene diamine measured using the new FANS-I configuration compared with that measured using the original configuration under identical beam collimations and measurement times. This gain in intensity is accompanied by a somewhat improved signal/noise ratio.

The enhanced capabilities of the FANS-I instrument have already been demonstrated for a variety of materials including protonic conductors, organic solids, metal hydrides, carbon nanotubes, and metal oxides. These experiments confirmed substantial reductions in required sample size and/or measurement time. For exam-

ple, Fig. 4 displays the temperature dependence of the NV spectrum for $\text{RbH}(\text{SO}_4)_{0.81}(\text{SeO}_4)_{0.19}$, a protonic conductor oxide with lattice protons that become mobile at temperatures less than 473 K. Vibrational spectra were collected with FANS-I at nine different temperatures in less than one day, a feat not possible using the original instrument. It proved particularly interesting that the energy of the mode near 100 meV decreases while that of the mode near 83 meV increases with increases in temperature concomitant with rapid decreases in both peak intensities. Indeed, these proton-related features largely disappear at temperatures as low as 200 K, indicating that there is significant proton motion even at this low temperature.

Figure 5 shows the FANS-I spectrum for solid 3-nitrophenol $[\text{HO}(\text{C}_6\text{H}_4)\text{NO}_2]$ compared with a GAUSSIAN calculation for the isolated molecule. This spectrum exemplifies the overall quality and high-resolution capabilities enabled by the marked improvement in sensitivity. There is fair agreement between calculation and experiment for many features in the vibrational density of states, although it is clear that significant intermolecular hydrogen bonding interactions in the solid cause strong perturbations in the OH wagging mode predicted near 45 meV for the isolated molecule.

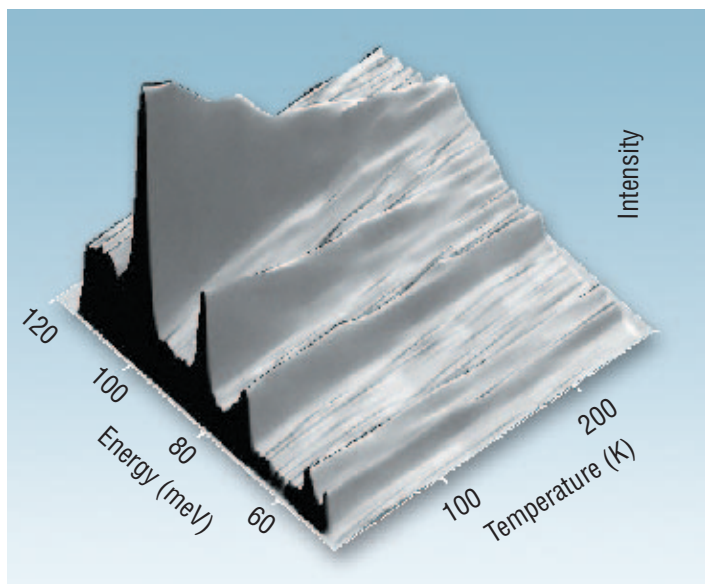


FIGURE 4. Temperature dependence of the NV spectrum of $\text{RbH}(\text{SO}_4)_{0.81}(\text{SeO}_4)_{0.19}$. (Data collected by T. Yildirim, NCNR.)

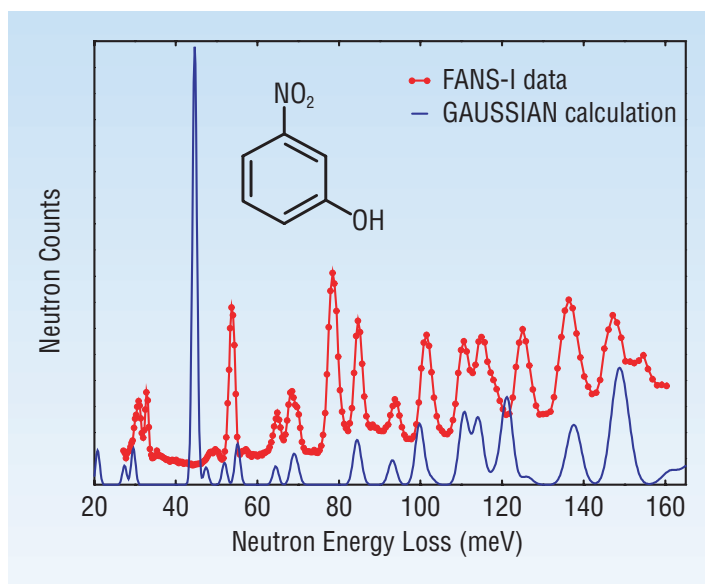


FIGURE 5. NV spectrum for solid 3-nitrophenol $[\text{HO}(\text{C}_6\text{H}_4)\text{NO}_2]$ at 10 K compared with a GAUSSIAN calculation (b3lyp/6-316*) for the isolated molecule. (Data collected by T. Heilweil, NIST Physics Laboratory, and C. M. Brown, U. Md. and NCNR.)